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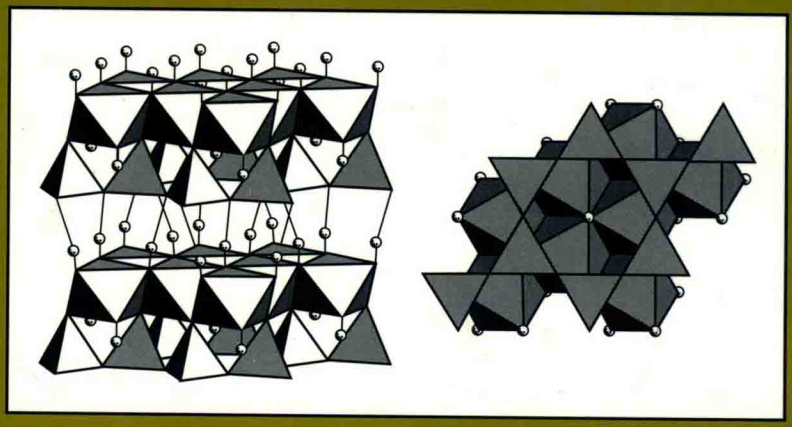
COLLOID AND SURFACE PROPERTIES OF CLAYS AND RELATED MINERALS



Rossman F. Giese
Carel J. van Oss

COLLOID AND SURFACE PROPERTIES OF CLAYS AND RELATED MINERALS

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Preface

Much has happened in the field of interfacial interactions among clay and other mineral particles surfaces since the publication of van Olphen's book in 1977 (see also van Olphen and Fripiat (1979)). Up to the middle 1980s one still habitually only considered van der Waals attractions and electrostatic repulsions as acting between such particles, even when immersed in a polar liquid such as water. The stability of aqueous suspensions of low electric charge particles was loosely ascribed to "steric" interactions, which however cannot be quantitatively expressed, e.g., in SI units. Even among the three kinds of van der Waals forces: dispersion (fluctuating dipole induced dipole), induction (dipole induced dipole) and orientation (dipole dipole) interactions, respectively alluded to as van der Waals-London, or dispersion forces, van der Waals-Debye, and van der Waals-Keesom forces, it remained difficult to decide which of these interactions was genuinely apolar. It was therefore even more uncertain how to make a sharp delineation between apolar forces (usually alluding to dipersion forces only) and polar forces (including hydrogen-bonding interactions). It was Dr. Manoj Chaudhury (1984), who first applied Lifshitzs' theory to macroscopic-scale colloidal interactions, which allowed him to establish for the first time a clear-cut distinction between apolar, or Lifshitz-van der Waals (LW) and polar, or electron-acceptor/electron-donor or Lewis acid-base (AB) interactions. Once this distinction had become obvious, it fairly quickly permitted the elaboration of a surface-thermodynamic approach comprising both apolar (LW) and polar (AB) interactions (van Oss *et al.*, 1987) and also includes electrostatic (EL) interactions (van Oss *et al.*, 1988).

Whilst for EL contributions the measurement methodology is an electrokinetic one (usually via micro-electrophoresis), for LW and AB interactions together the method of choice remains the determination of contact angles with drops of a small number of appropriate, relatively high-energy liquids,

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apolar (e.g., diiodomethane) as well as polar (water, glycerol, formamide), deposited on a solid surface. This still remains the preferred approach, as it is the only one that allows the analysis of the surface properties of condensed-phase materials at their exact surface (not 1, 5, 10 or more nm below it). This is particularly important because, e.g., solid and liquid materials interact with one another precisely via their exact surfaces. Contact angle measurements are of course easy to perform on, e.g., flat surfaces of large mineral crystals. With small clay or other mineral particles, however, this is a somewhat different matter. When one has particles of a swelling clay (e.g., smectite particles) it is relatively easy to deposit such clay particles by sedimentation from an aqueous suspension onto a smooth surface of plastic or glass, whereupon after drying one obtains perfectly smooth surfaces (Giese *et al.*, 1990; van Oss *et al.*, 1990) with which contact angle measurements are quite easily performed. A difficulty arises however with non-swelling clays and with practically all other mineral particles. These can of course quite easily be deposited on a smooth flat surface but by their very granular nature they form, upon drying, a rough surface, usually with a radius of curvature of roughness of 1 or more μm. Unfortunately, surfaces exhibiting a degree of roughness greater than 1 μm, give rise to strongly exaggerated contact angles, so that direct contact angle analysis cannot be used in these cases (Chaudhury, 1984). Again, encouraged by Chaudhury (personal communication, ca. 1989), the thin layer wicking technique was developed by a number of us (Costanzo *et al.*, 1991; van Oss *et al.*, 1992), with the help of Karen Murphy and Drs. Janice Norris, Z. Li and Wenju Wu. This approach made it possible for the first time to measure the surface properties of many of the particles listed in Chapter 9. Only swelling clays allow the determination of their surface properties by direct contact angle measurement (see also Chapter 9), but wicking cannot be done with swelling clays because during the capillary rise measurements, involved in wicking, the liquid migration which should occur in one direction only, is severely diminished in that one direction, when liquid uptake occurs at the same time in two other directions, due to swelling.

There was hitherto no absolute certainty that contact angle measurements via contact angles and thin layer wicking really afforded measurements of the same contact angle values. However, it was subsequently found that the use of non-swelling monosized cubical synthetic hematite (Fe_2O_3) particles of about 0.6 μm, permitted the deposition of exceedingly smooth layers of particles onto glass surfaces. These could be utilized for thin layer wicking

as well as for direct contact angle measurements and thus furnished the experimental proof that the same contact angles would result from either technique (Costanzo *et al.*, 1995).

Interfacial, non-covalent interaction energies between various identical, or different, condensed-phase materials, taking place in water, are here alluded to as ΔG^{IF}_{1w1} and ΔG^{IF}_{1w2}, respectively (see Tables 1 and 2). These represent either hydrophobic attractions (when $\Delta G^{IF} < 0$) or hydrophilic repulsions (when $\Delta G^{IF} > 0$). Hydrophobic attractions are often alluded to as caused by the "Hydrophobic Effect", whilst hydrophilic repulsions correspond to what has been called "Hydration Pressure". Both free energies of hydrophobic attraction and hydrophilic repulsion can be obtained from the measurements of surface properties of condensed-phase materials, and expressed quantitatively in SI units. Both types of interaction are relatively long-range: their interaction energies decay as a function of distance, ℓ, as $\exp[(\ell_o - \ell)/\lambda]$, where ℓ_o is the minimum equilibrium distance between two surfaces ($\ell_o = 0.157$ nm), and λ is the characteristic length for water ($\lambda \approx 1.0$ nm). Hydrophobic attraction is due to the Lewis acid-base (AB) free energy of *cohesion* between the water molecules (w), whereas hydrophilic repulsion is a consequence of the (AB) free energy of *attraction* between the water molecules (w) electron-acceptor sites (i.e., their H-atoms) and the electron-donor sites on the immersed hydrophilic materials or molecules (1 and/or 2). When the free energy of hydrophilic repulsion is greater than the underlying free energy of hydrophobic attraction (the latter, by its very nature is always present), a net repulsion ensues. Compared with AB interactions, occurring in water, Lifshitz-van der Waals (LW) interactions provide only a minor contribution to the interfacial free energies, ΔG^{IF}_{1w1} or ΔG^{IF}_{1w2}, as do electrostatic interactions (EL), which however for practical reasons are considered separately from interfacial (IF) interactions.

Most of the data generated on the surface properties of clays and other mineral particles, in the decade from 1988 to 1998 are discussed in the various Chapters of this book, especially in Chapter 9. A summary of some of the more important general applications of the above-described approach, treating polar interactions among clays and other mineral particles in aqueous media, is outlined in Tables 1 and 2.

The material presented in this book is designed to be of use to a wide variety of people, often with very different backgrounds. In order to provide a reasonably self contained volume, there are a number of sections which are

Phenomenon or Method	Mechanism	Utilizations	Chapter-Section
Stability of aqueous particle suspensions, as determined via the extended DLVO approach	Hydrophobic (AB) repulsion vs. LW attraction; see equations in Table 8.4	Determination of suspension stability, e.g., for the prediction of movements of soils, or of volcanic ash deposits; flocculation	8.8, 8.9
Connection between ΔG^{IF}_{1w1} of 1 with respect to solvent (e.g., water) and the solubility of 1	see Eqn. (8.7)	Computation of solubility, or of critical micelle concentration from ΔG^{IF}_{1w1}, or vice-versa	8.10
Swelling of clay minerals	Hydrophilic (AB) repulsion	formation of impermeable environmental barriers for containment of toxic or radioactive wastes	8.13, 8.15

Table 1: *Applications of quantitatively expressed ΔG^{IF}_{1w1} values, i.e., the free energies of interfacial, non-covalent interactions between identical particles or molecules, 1, immersed in water, w.*

Phenomenon or Method	Mechanisms	Utilizations	Chapter-Section
Adsorption and adhesion	Hydrophobic (AB) attraction; for dissociation, followed by hydrophilic (AB) repulsion	Liquid chromatography methods (e.g., reversed phase liquid chromatography), other separation and purification methods; removal of toxic spills; hydrocarbon recovery, various utilizations of zeolites; additives of clays in the paper industry; flotation	2, 8.11, 8.12,10

Table 2: *Applications of quantitatively expressed ΔG^{IF}_{1w2} values, i.e., the free energies of interfacial, non-covalent interactions between different particles and/or molecules, 1 and 2, immersed in water, w.*

intended as background material. For example, Chapters 3 and 4 provide an introduction to clay mineralogy and common minerals in general. Clearly, for a reader with a background in mineralogy, these chapters will be of marginal interest if any. The book also provides a summary of modern colloid chemistry (Chapters 5–8). More detail can be found in the book by van Oss (van Oss, 1994). The surface thermodynamic data on minerals are presented in Chapter 9. Clearly, not all minerals have been examined. The study of the surface chemistry of minerals is still young, so there are a number of minerals and mineral groups which have not yet been examined. It is the hope of the authors that the present volume will stimulate others to expand our knowledge.

The pictorial representations of mineral structures were created using the software package ATOMS, version 3.2 by Eric Dowty. P. Avery prepared many of the mineral samples for the measurements reported in Chapter 9.

Rossman F. Giese

Carel J. van Oss

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